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Static magnetization induced by time-periodic fields with zero mean

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Abstract

We consider a single spin in a constant magnetic field or an anisotropy field. We show that additional external time-periodic fields with zero mean may generate nonzero time-averaged spin components which vanish for the time-averaged Hamiltonian. The reason is a lowering of the dynamical symmetry of the system. A harmonic signal with proper orientation is enough to display the effect. We analyze the problem both with and without dissipation. The results are of importance for controlling the system's state using high- or low-frequency fields and for using new resonance techniques which probe internal system parameters, to name a few. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Usually nonzero averages of observables, which would be expected to be zero by symmetry considerations, are generated either by permanent symmetry breaking external fields, or by internal interactions which may lead to phase transitions. However as we will show below, such a situation is also possible if we use time-periodic fields with zero mean. The general idea behind the following results is purely symmetry related, and thus it seems to be worthwhile to understand the mechanisms which may lead to nonzero averages if such fields are applied. This work is motivated by a recent paper [1] where similar ideas have been used to explain the phenomenon of directed

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currents in driven systems. The essence of the present paper is that we can lower the symmetry of a given dynamical system by applying time-periodic fields with zero mean, i.e., that the time-averaged Hamiltonian displays symmetries which would imply zero averages for corresponding observables. It will be the symmetry breaking in the temporal evolution which induces nonzero averages.

Let us start our considerations with a model describing an $s = \frac{1}{2}$ spin in a constant field h_z directed along the z-direction and a time-periodic field $h_x(t)$ with period T and zero mean directed along the x-direction. The Hamiltonian is given by

$$H = h_z S_z + h_x(t) S_x \tag{1}$$

(here $S_{x,y,z}$ are the spin component operators related to the corresponding Pauli matrices, e.g. [2]). For the moment we assume that $|h_x(t)| \ll 1$ and the frequency $\omega = 2\pi/T \ll 1$. In that case, we can use the adiabatic approximation and neglect Zener transitions. The two eigenvalues of H for a given value of h_x are

$$\lambda_\pm = \pm \tfrac{1}{2}\sqrt{h_z^2 + h_x^2}\ . \tag{2}$$

The expectation value for S_x in these states is given by

$$\langle S_x \rangle = \frac{h_x}{2\sqrt{h_z^2 + h_x^2}}\ . \tag{3}$$

Now, we assume that the spin is in any of the two states. Slow variation of h_x in time will keep the system in that state. Let us average $\langle S_x \rangle$ over one period of oscillation. Because $\langle S_x \rangle$ is odd in h_x, we will obtain nonzero time averages for the x-component of the spin if e.g. $\int_0^T h_x^3 \, \mathrm{d}t \neq 0$. This is possible if $h_x(t)$ contains several harmonics (SH), e.g.

$$h_x(t) = h_1 \cos(\omega t) + h_2 \cos(2\omega t + \xi) \tag{4}$$

(see also [1]). In that case in lowest order in h_1, h_2 we obtain

$$\langle S_x \rangle = -\frac{3}{16} \frac{h_1^2 h_2}{h_z^3} \cos \xi\ . \tag{5}$$

We conclude this example by stating that it is possible to generate a nonzero average S_x spin component by applying a permanent field in the z-direction and a time-periodic field with SH and zero average in the x-direction.

2. Density matrix approach

Let us relate the results from the example given above to symmetry considerations. The Hamiltonian H should be a periodic function of time $H(t) = H(t + T)$. Instead of solving the time-dependent Schrödinger equation which would bring us to the analysis of unitary Floquet matrices [3,4], we follow the density matrix approach which is suitable since we want to average over different initial conditions and are thus facing the dynamics of mixed states. We assume that the density matrix ρ satisfies the quantum Liouville equation [2] with a linear relaxation term [5]

$$\frac{\partial \rho}{\partial t} = \mathrm{i}[H, \rho] - v(\rho - \rho_\beta) , \tag{6}$$

where $[A, B] = AB - BA$, ρ_β is some equilibrium density matrix parametrized by the inverse temperature β and v is a phenomenological parameter measuring the coupling strength of the system described by H to some environment. Note that v is the characteristic inverse relaxation time of H in the environmental bath.

Let us further define

$$H_0 = \frac{1}{T}\int_0^T H(t)\,\mathrm{d}t \quad \text{and} \quad H_1(t) \equiv H(t) - H_0 . \tag{7}$$

Note that $\int_0^T H_1(t)\,\mathrm{d}t = 0$. Then we may choose

$$\rho_\beta = \frac{1}{Z}\,\mathrm{e}^{-\beta H_0} \quad \text{with } Z = \mathrm{Tr}(\mathrm{e}^{-\beta H_0}) . \tag{8}$$

We define the value $\bar{A}(t)$ of an observable characterized by the operator A as

$$\bar{A}(t) = \mathrm{Tr}(A\rho(t)) . \tag{9}$$

The time average of $\bar{A}(t)$ shall be defined as

$$\tilde{A} = \lim_{t' \to \infty} \frac{1}{t'} \int_0^{t'} \bar{A}(t)\,\mathrm{d}t . \tag{10}$$

The averaged attenuation power (the rate of energy transfer from the time-periodic field to the heat bath) is given by $W = v(\tilde{H}_0 - \mathrm{Tr}(H_0\rho_\beta))$.

We chose the relaxation term in (6) in an oversimplified form. There are many theories which exploit different concrete relaxation mechanisms (e.g. [6] and references therein). The reason for choosing (6) instead is that it allows to discuss the following symmetry breaking without entering the details of the concrete dissipation mechanism. In other words, we deliberately choose the simplest dissipation term which conserves all symmetries of our dynamical system except time reversal.

Eq. (6) is a linear equation for the matrix coefficients of ρ with inhomogeneous terms due to ρ_β. The general solution is given by a sum of the general solution of the homogeneous equation (put $\rho_\beta = 0$ in (6)) and a particular solution of the full equation. Since the homogeneous solution for $v = 0$ is given by some unitary time evolution, $v > 0$ will cause all solutions of the homogeneous equation to decay to zero for infinite time. For $t \gg 1/v$, any particular solution of the inhomogeneous equation tends to a unique time-periodic solution – the attractor of (6). This allows us to choose any (reasonable) initial condition $\rho(t=0)$. If H, $\rho(t=0)$ and ρ_β are invariant under certain unitary transformations, it immediately follows that $\rho(t)$ keeps those symmetries, and consequently the attractor will have the same symmetries too. For large temperatures, ρ_β approaches the unity matrix (up to some factor). Consequently in that limit, whatever is the time dependence of $H(t)$, the solution of (6) will approach ρ_β. Finally, we note that due to $\mathrm{Tr}\,\rho_\beta = 1$ any choice of $\rho(t=0)$ with $\mathrm{Tr}\,\rho(t=0) = 1$ implies $\mathrm{Tr}\,\rho(t) = 1$ for all t.

2.1. The $s = \frac{1}{2}$ case

Let us consider (6) for

$$H = h_0 S_z + h(t)(\alpha S_x + \gamma S_z)\,, \tag{11}$$

where $\alpha = \sin(\phi)$ and $\gamma = \cos(\phi)$. This model describes a spin in a constant magnetic field pointing in the z-direction, under the influence of an additional time-periodic field $h(t) = h(t+T)$. This oscillating field should have a zero mean: $\int_0^T h(t)\,\mathrm{d}t = 0$. Let us define $h(t)$ having T_a symmetry if

$$h(t) = -h(-t) \equiv h_a(t)\,, \tag{12}$$

T_s symmetry if

$$h(t) = h(-t) \equiv h_s(t) \tag{13}$$

and T_{sh} symmetry if

$$h(t) = -h(t + T/2) \equiv h_{sh}(t) \tag{14}$$

(note that in the first two cases any argument shift is allowed, so that e.g. $h(t) = \cos(t+\mu)$ possesses all three symmetries). For a monochromatic field (MCF) $h(t)$ and $\phi = \pi/2$ (11) is the classical setup for performing magnetic resonance (MR) experiments [7,8].[1]

For the $s = \frac{1}{2}$ case, the spin component operators are given by the Pauli matrices: $S_{x,y,z} = \frac{1}{2}\sigma_{x,y,z}$. The density matrix ρ has three independent real variables. Using the variables $\bar{S}_{x,y,z}$ we find

$$\dot{\bar{S}}_x = (h_0 + \gamma h(t))\bar{S}_y - v\bar{S}_x\,, \tag{15}$$

$$\dot{\bar{S}}_y = \alpha h(t)\bar{S}_z - (h_0 + \gamma h(t))\bar{S}_x - v\bar{S}_y\,, \tag{16}$$

$$\dot{\bar{S}}_z = -\alpha h(t)\bar{S}_y - v(\bar{S}_z - C)\,, \tag{17}$$

where $C = \frac{1}{2}\tanh(h_0\beta/2)$. Note that the obtained set of equations for $v = 0$ is equivalent to the Heisenberg equations for the operators $S_{x,y,z}$ and thus also to the equations of motion for a classical spin. In fact, (15)–(17) is a particular case of the phenomenological Bloch equations [7][2] which are well known in the theory of nuclear magnetic resonance.

Let us discuss the symmetries of (15)–(17) which conserve H_0, i.e., $\bar{S}_z \to S_z$. Consider the case $\gamma = 0$: if $h(t) \equiv h_{sh}(t)$ then a symmetry operation Q_1 is

$$\bar{S}_x \to -\bar{S}_x, \quad \bar{S}_y \to -\bar{S}_y, \quad \bar{S}_z \to \bar{S}_z, \quad t \to t + \frac{T}{2}\,. \tag{18}$$

[1] Most of the theoretical studies also confine to $\gamma = 0$. For an exception see Ref. [9].

[2] Our equations (15)–(17) differ from the corresponding Bloch equations since we did not choose different relaxation times for the different spin components. However, choosing different relaxation times would not change the symmetry properties discussed in the paper, and consequently a variation of these relaxation times would cause only a quantitative change.

If Q_1 holds, we conclude that $\tilde{S}_x = \tilde{S}_y = 0$, while $\tilde{S}_z$ may be nonzero. Consider $\gamma = 0$ and $v = 0$: if $h(t) \equiv h_a(t)$ then a symmetry operation Q_2 is

$$\bar{S}_x \to -\bar{S}_x, \quad \bar{S}_y \to \bar{S}_y, \quad \bar{S}_z \to \bar{S}_z, \quad t \to -t\,. \tag{19}$$

If Q_2 holds it follows that $\tilde{S}_x = 0$, while $\tilde{S}_{y,z}$ may be nonzero. Finally for $v = 0$ and $h(t) \equiv h_s(t)$ a symmetry operation Q_3 is

$$\bar{S}_x \to \bar{S}_x, \quad \bar{S}_y \to -\bar{S}_y, \quad \bar{S}_z \to \bar{S}_z, \quad t \to -t\,. \tag{20}$$

If Q_3 holds it follows that $\tilde{S}_y = 0$, while $\tilde{S}_{x,z}$ may be nonzero.

Let us note some consequences. If we choose $h(t) = h_1 \cos(\omega t)$, then the classical MR setup with $\gamma = 0$ (Q_1) yields nonzero values for $\tilde{S}_z$ only [7,8]. If the probing field is not perpendicular to the z-axis ($\gamma \neq 0$), nonzero values appear for $\tilde{S}_x$ and $\tilde{S}_y$ as well. $\tilde{S}_y$ will vanish in the limit of zero coupling to the environment $v \to 0$ (Q_3), so that this average can be used to measure the coupling strength. Applying, e.g. $h(t) = h_1 \sin(\omega t) + h_2 \sin(2\omega t)$ (having h_a symmetry but not h_{sh} and h_s one), we can suppress the value of $\tilde{S}_x$ relatively to $\tilde{S}_y$ for $\gamma \to 0$ and $v \to 0$ keeping $\tilde{S}_y$ finite (Q_2)!

Analytical solutions to (15)–(17) can be found e.g. for large $v \gg 1$. For that we rewrite Eqs. (15)–(17) in the following way:

$$\bar{S}_x = \frac{1}{v}[-\dot{\bar{S}}_x + (h_0 + \gamma h(t))\bar{S}_y]\,, \tag{21}$$

$$\bar{S}_y = \frac{1}{v}[-\dot{\bar{S}}_y + \alpha h(t)\bar{S}_z - (h_0 + \gamma h(t))\bar{S}_x]\,, \tag{22}$$

$$\bar{S}_z = C + \frac{1}{v}[-\dot{\bar{S}}_z - \alpha h(t)\bar{S}_y]\,. \tag{23}$$

In 0th order in $1/v$, we have $\bar{S}_x = \bar{S}_y = 0$ and $\bar{S}_z = C$. Inserting this solution into the right-hand sides of Eqs. (21)–(23) we obtain the solution in first order in $1/v$. Continuing to do so, i.e., expanding in $1/v$ and in addition averaging over time, we find in lowest orders,

$$\begin{aligned}\tilde{S}_x &= C\alpha\gamma\langle h^2\rangle\frac{1}{v^2} - C\alpha(-\gamma\langle h\ddot{h}\rangle + 3\gamma h_0^2\langle h^2\rangle \\ &\quad + (\alpha^2 + 3\gamma^2)h_0\langle h^3\rangle + \gamma(\gamma^2+\alpha^2)\langle h^4\rangle)\frac{1}{v^4} + O\left(\frac{1}{v^5}\right)\,,\end{aligned} \tag{24}$$

$$\tilde{S}_y = -C\alpha[2\gamma h_0\langle h^2\rangle + (\gamma^2+\alpha^2)\langle h^3\rangle]\frac{1}{v^3} + O\left(\frac{1}{v^5}\right)\,, \tag{25}$$

where $\langle f(t)\rangle = \frac{1}{T}\int_0^T f(t)\,\mathrm{d}t$. It is easy to cross check that all symmetry statements from above are correct. Nonzero values for $\langle h^3\rangle$ can be obtained e.g. with $h(t) = h_1 \sin(\omega t) + h_2 \sin(2\omega t + \xi)$ for $\xi \neq 0, \pi$ (see also [1]).

In Fig. 1 we show the dependence of $\tilde{S}_{x,y,z}$ on ω for $h(t) = \sqrt{2}\cos\omega t$, $\phi = \pi/4$, $h_0 = 3$, $v = 0.1$ and $\beta = 10$. The time-periodic field has a large amplitude compared to typical MR setups [7,8]. This causes the $\tilde{S}_z$ curve to show a rather broad peak at $\omega \approx h_0$ – the

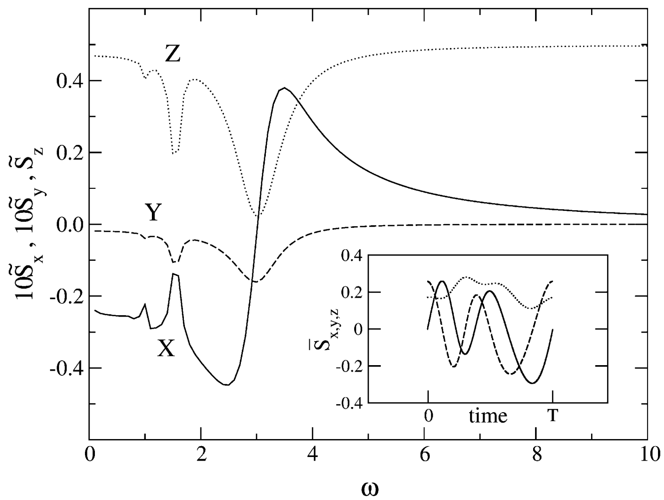

Fig. 1. $10\tilde{S}_x$ (solid), $10\tilde{S}_y$ (dashed) and $\tilde{S}_z$ (dotted) as functions of ω (see text for parameters). Inset: $\bar{S}_{x,y,z}$ versus time for one period of $h(t)$ at $\omega = 1.5$ (same line codes as in Fig. 1). Note that functions are not scaled here!

position of the expected MR resonance. We also observe subharmonic peaks at lower frequencies which are due to the nonlinear response induced by the large amplitude of the driving field. The main observation is the presence of nonzero values for $\tilde{S}_{x,y}$ (for convenience, these averages are scaled by a factor of 10 in Fig. 1). The dependence of $\tilde{S}_x$ and $\tilde{S}_y$ on ω shows rather complex structures. We find typically that the dependence of these averages on ω becomes oscillatory for small $\omega \ll h_0$, whereas large ω values yield smooth decay curves. It is also important to notice that the fluctuations of $\bar{S}_x$ and $\bar{S}_y$ around their mean values may happen with amplitudes being one order of magnitude larger than the mean values (see inset in Fig. 1).

2.2. The $s = 1$ case

The above results hold also for larger spins. To show that they also hold for internal anisotropy fields rather than external fields, we consider a spin with $s = 1$ and the Hamiltonian

$$H = S_z^2 + h(t)(\alpha S_x + \gamma S_z)\,, \tag{26}$$

which describes a spin with an anisotropy along the z-axis (S_z^2) under the influence of an external magnetic field $h(t)$ parallel to the xz-plane. The magnetic field is again time-periodic with period T and has a zero mean. The 3×3 Hermitian density matrix ρ has 8 independent real parameters. Since H in (26) is a real symmetric matrix, we can define $\rho = R + \mathrm{i}I$ where R is a real symmetric matrix and I a real antisymmetric one. Noting that ρ_β also is a real diagonal matrix, (6) can be rewritten as

$$\frac{\partial R}{\partial t} = -[H, I] - \nu(R - \rho_\beta)\,, \tag{27}$$

$$\frac{\partial I}{\partial t} = [H,R] - \nu I \,. \tag{28}$$

It follows[3] that

$$\bar{S}_x = \sqrt{2}(R_{12} + R_{23}), \qquad \bar{S}_y = -\sqrt{2}(I_{12} + I_{23}) \quad \text{and} \quad \bar{S}_z = R_{11} - R_{33} \,. \tag{29}$$

Using the abbreviations,

$$P_x = \sqrt{2}(R_{12} - R_{23}), \quad P_y = \sqrt{2}(I_{12} - I_{23}), \quad P_z = R_{11} + R_{33} \,,$$

$$\hat{R}_{13} = \sqrt{2}R_{13}, \quad \hat{I}_{13} = \sqrt{2}I_{13}, \quad \hat{R}_{22} = \sqrt{2}R_{22} \,,$$

$$D^{-1} = 1 + 2\mathrm{e}^{-\beta} \quad \text{and} \quad F^{-1} = 2 + \mathrm{e}^{\beta} \,,$$

the equations of motion become

$$\begin{aligned}
\dot{\bar{S}}_x &= -P_y + \gamma h \bar{S}_y - \nu \bar{S}_x \,, \\
\dot{P}_x &= \bar{S}_y - \gamma h P_y + \sqrt{2}\alpha h \hat{I}_{13} - \nu P_x \,, \\
\dot{\bar{S}}_y &= -P_x - \gamma h \bar{S}_x + \alpha h \bar{S}_z - \nu \bar{S}_y \,, \\
\dot{P}_y &= \bar{S}_x + \gamma h P_x + \alpha h[\sqrt{2}\hat{R}_{22} - P_z - \sqrt{2}\hat{R}_{13}] - \nu P_y \,, \\
\dot{\bar{S}}_z &= \alpha h \bar{S}_y - \nu \bar{S}_z \,, \\
\dot{P}_z &= \alpha h P_y - \nu[P_z - 2F] \,, \\
\dot{\hat{R}}_{13} &= -2\gamma h \hat{I}_{13} + \sqrt{2}\alpha h P_y - \nu \hat{R}_{13} \,, \\
\dot{\hat{I}}_{13} &= 2\gamma h \hat{R}_{13} - \sqrt{2}\alpha h P_x - \nu \hat{I}_{13} \,, \\
\dot{\hat{R}}_{22} &= \sqrt{2}\alpha h P_y - \nu[\hat{R}_{22} - \sqrt{2}D] \,.
\end{aligned} \tag{30}$$

These equations conserve the trace $\mathrm{Tr}\,\rho \equiv P_z + \hat{R}_{22}/\sqrt{2} = 1$.

Now, we can discuss the symmetries of (30) which change the sign of $\bar{S}$. Two of them hold only for $\nu = 0$. First, if $h(t) \equiv h_a(t)$, then the equations are invariant under change of sign of the variables t, $\bar{S}_x$, $\bar{S}_y$, $\bar{S}_z$ (leaving all other variables unchanged). A second case takes place if $h(t) \equiv h_s(t)$. Then changing the sign of t, $\bar{S}_y$, P_y and $\hat{I}_{13}$ (leaving all other variables unchanged) is an operation which keeps Eqs. (30) invariant. These two cases imply that if $h(t)$ is antisymmetric, then for vanishing dissipation $\nu \to 0$, $\tilde{S}_{x,y,z} \to 0$, while for symmetric $h(t)$ the same limit provides a vanishing of the y-component only, $\tilde{S}_y \to 0$.

For the general case $\nu \neq 0$, two more symmetries may take place. If $\gamma = 0$ (the field $h(t)$ acts perpendicularly to the anisotropy z-axis), changing the sign of $\bar{S}_y, \bar{S}_z, P_x$ and $\hat{I}_{13}$ (and keeping all others) leaves (30) invariant. Finally if $h(t) \equiv h_{sh}(t)$, the shift $t \to t + T/2$ and simultaneous change of sign of the variables $\bar{S}_x$, $\bar{S}_z$, P_y, $\hat{I}_{13}$ do not

[3] The matrix representations for $S_{x,y,z}$ are taken from Ref. [10].

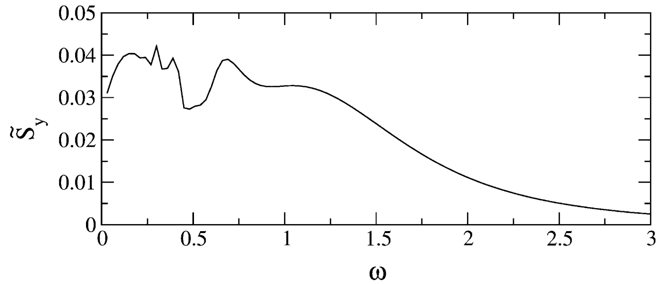

Fig. 2. $\tilde{S}_y$ as a function of ω (see text for parameters).

change the equations. It follows that $\tilde{S}_y = \tilde{S}_z = 0$ for $\gamma = 0$ and $\tilde{S}_x = \tilde{S}_z = 0$ for $h(t)$ having shift symmetry.

It is interesting to note that for a MCF, $h(t) = \cos \omega t$ and $v \neq 0$, $\gamma \neq 0$, the spin will point on average in the y-direction, i.e., perpendicular to the plane spanned by the driving field and the local anisotropy axis. In Fig. 2 we plot the dependence of $\tilde{S}_y$ on ω for this case ($\beta = 10$, $v = 0.1$, $\gamma = \alpha = 1$), which confirms the symmetry considerations. Note that $\tilde{S}_x$ and $\tilde{S}_z$ are less than 10^{-8} as found in the numerical studies.

To conclude this case, we remark that it is again an easy task to perform expansions in $1/v$ for large v values as shown above for the $s = \frac{1}{2}$ case. The resulting expressions also confirm the symmetry considerations.

3. Discussions and summary

It is interesting to note that similar models have been used to study the issue of large spin tunneling in the presence of ac magnetic fields, e.g. in Ref. [11], a model $H = -\gamma S_z^2 - \alpha[\cos(\omega t) S_x + \sin(\omega t) S_y]$ is discussed. It is easy to check that whereas for $\alpha = 0$, the equations of motion are invariant under a sign change of S_z, this symmetry is lost for nonzero α. Consequently the ac fields aligned in the xy-plane will induce a magnetic moment with nonzero z-component.

Let us estimate the expected induced fields for the case of electron paramagnetic resonance (see e.g. Ref. [8]) with paramagnetic centers having $s = \frac{1}{2}$ at temperature $T = 1$ K.[4] For a center concentration of 10^{22} cm^{-3}, we may neglect effects of interaction. A constant field $h_0 = 0.15$ T leads to a resonance frequency of 2 GHz. The difference from standard MR setups is that we choose the angle between the permanent and the driving field to be 45°, and the driving field being only twice weaker than the permanent field, i.e., 0.075 T. Taking the inverse relaxation times to be of the order of 0.1 GHz, Eqs. (15)–(17) predict a dimensionless value of $\tilde{S}_y \sim 0.002$. This implies that the induced magnetic field component perpendicular to the two applied fields will

[4] We did not choose nuclear spins because their magnetic moments are 1000 times less than the ones of electrons. However we do not want to exclude that the proposed effects may also be detected in nuclear magnetic resonance setups.

be of the order of 2 G. The fact that the permanent and ac fields have comparable amplitudes causes a smearing out of the original resonance signal. Consequently, it increases the range of frequencies (around the original resonance frequency) where the induced magnetic fields may be observed.

One may enhance the expected effect further by increasing the concentration of paramagnetic centers. This calls for a study of the role of interactions. An analysis of two spins with exchange interaction shows that ferromagnetic interaction will increase the induced field, whereas strong antiferromagnetic interaction will decrease the induced field [12]. Note that the above example is just one possibility of many realizations which will be discussed in detail elsewhere.

Let us summarize the results presented. We have shown that time-periodic magnetic fields with zero mean may induce nonzero averages of spin components which would be strictly zero in the absence of these fields. The spin simultaneously experiences some local anisotropy field or simply an external constant field. In addition, the spin is coupled to some thermal environment characterized by some finite temperature and a characteristic relaxation time. [5] The reasoning follows symmetry considerations of the dynamical equations. In the quantum case, we solve the (purely linear!) equations of motion for the independent components of the density matrix. Remarkably, the symmetry properties obtained from both the approaches coincide. For the spin $\frac{1}{2}$ case, we propose a MR experiment to observe the effect. The main difference from standard MR setups is that the static and ac fields are not perpendicular to each other and that the ac field has amplitude comparable or slightly less than the one of the static field.

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[5] A number of studies (e.g. Ref. [13]) suggest that the dissipation in (6) may include retardation effects. Evidently these terms will not change the considered symmetries.